

Density Functionals in the Presence of Magnetic Field

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Abstract

In this paper density functionals for Coulomb systems subjected to electric and magnetic fields are developed. The density functionals depend on the particle density, ρ , and paramagnetic current density, j^p . This approach is motivated by an adapted version of the Vignale and Rasolt formulation of Current Density Functional Theory (CDFT), which establishes a one-to-one correspondence between the non-degenerate ground-state and the particle and paramagnetic current density. Definition of N -representable density pairs (ρ, j^p) is given and it is proven that the set of v -representable densities constitutes a proper subset of the set of N -representable densities. For a Levy-Lieb type functional $Q(\rho, j^p)$, it is demonstrated that (i) it is a proper extension of the universal Hohenberg-Kohn functional, $F_{HK}(\rho, j^p)$, to N -representable densities, (ii) there exists a wavefunction ψ_0 such that $Q(\rho, j^p) = (\psi_0, H_0 \psi_0)_{L^2}$, where H_0 is the Hamiltonian without external potential terms, and (iii) it is not convex. Furthermore, a convex and universal functional $F(\rho, j^p)$ is studied and proven to be equal the convex envelope of $Q(\rho, j^p)$. For both Q and F , we give upper and lower bounds.

I. INTRODUCTION

The theoretical foundation of Density Functional Theory (DFT) is the Hohenberg-Kohn theorem [1] that states that the particle density of a quantum mechanical system determines the scalar potential up to a constant. Arguments have been put forward that this theorem could be generalized to include systems with magnetic fields [2–4]. These arguments rely on either the paramagnetic current density or the total current density being used together with the particle density to determine the scalar potential and vector potential of the system. Nonetheless, for the formulation with paramagnetic current density, counterexamples have been constructed that exclude the existence of a Hohenberg-Kohn theorem for such a formulation (see for instance [5] where this was first demonstrated, or [6] for more mathematical details in the one-electron case). Moreover, the existence of a Hohenberg-Kohn theorem for the formulation with the total current density is still an open question [6, 7], since the proofs of [3] and [4] do not hold. (The error in [3] was highlighted in [6] and the error in [4] was pointed out in [7].)

However, in an adapted version of the Vignale and Rasolt formulation of Current Density Functional Theory (CDFT), the particle density and the paramagnetic current density determine the non-degenerate ground-state [2, 6]. This allows a Hohenberg-Kohn functional to be defined, from which other density functionals can be developed. Following Lieb’s programme for DFT [8], the issue of establishing a mathematically rigorous CDFT formulated with the paramagnetic current density will here be addressed.

The aims of this article are the following:

- (i) *Define the set of N -representable particle and paramagnetic current densities.* The definition is motivated by Proposition 3, and Proposition 4 shows that this set is convex.
- (ii) *For N -representable particle and paramagnetic current densities, study a Levy-Lieb type functional $Q(\rho, j^p)$.* In Theorem 5, $Q(\rho, j^p)$ is proven to be a proper extension of the universal Hohenberg-Kohn functional and, moreover, it is proven that there exists a minimizer such that $Q(\rho, j^p) = (\psi_0, H_0 \psi_0)_{L^2}$. Proposition 8 demonstrates that $Q(\rho, j^p)$ is not a convex functional, which motivates
- (iii) *Investigate a convex and universal particle and paramagnetic current density functional, $F(\rho, j^p)$.* In Theorem 11, it is proven that $F(\rho, j^p)$ equals the convex envelope of

$Q(\rho, j^p)$. Furthermore, in Theorem 13, the minimization of

$$F(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2)$$

is connected with a set of Euler-Lagrange equations.

(iv) *Give upper and lower bounds for particle and paramagnetic current density functionals.* Bounds for both $Q(\rho, j^p)$ and $F(\rho, j^p)$ are found in Theorem 14, Proposition 16 and Corollary 17. Proposition 16 and Corollary 17 require that the vorticity is zero.

II. PRELIMINARIES

We will in this paper consider a system of N interacting electrons. The Hamiltonian of the system is given by (in suitable units)

$$H(v, A) = \sum_{k=1}^N ((i\nabla_k - A(x_k))^2 + v(x_k)) + \sum_{1 \leq k < l \leq N} |x_k - x_l|^{-1}, \quad (1)$$

where $v(x)$ is the scalar potential and $A(x)$ the vector potential, with components $A^k(x)$ $k = 1, 2, 3$, such that $B(x) = \nabla \times A(x)$, where $B(x)$ is the magnetic field. The following will be assumed: (i) there is a lowest eigenvalue e_0 of $H(v, A)$ with $\dim \ker(e_0 - H) = 1$, (ii) the solution of $H(v, A)\psi = e_0\psi$ fulfils $\psi \neq 0$ almost everywhere (a.e.), and (iii) the magnetic field vanishes outside some large sphere (B has compact support) and we may take $A^k(x)$ to be bounded. See [6] for further discussion about assumptions (i) and (ii).

Some different function spaces will be used in the forthcoming discussion. A function f that satisfies $\int_{\mathbb{R}^n} |f|^p < \infty$, for some $p \in [1, \infty)$, is said to belong to the normed space $L^p(\mathbb{R}^n)$ with norm $\|f\|_{L^p(\mathbb{R}^n)} = (\int_{\mathbb{R}^n} |f|^p)^{1/p}$. For $R > 0$, let $B_R = \{x \in \mathbb{R}^n \mid |x| \leq R\}$. Then $f \in L^p_{\text{loc}}(\mathbb{R}^n)$ if for any B_R we have that $\|f\|_{L^p(B_R)} = \left(\int_{B_R} |f|^p\right)^{1/p} < \infty$. The normed space $L^\infty(\mathbb{R}^n)$ consists of those functions f that satisfy $\|f\|_{L^\infty(\mathbb{R}^n)} = \text{ess sup}\{|f| \mid x \in \mathbb{R}^n\} < \infty$. A function $f \in L^2(\mathbb{R}^n)$ that satisfies $\int_{\mathbb{R}^n} |\nabla f|^2 < \infty$ belongs to $H^1(\mathbb{R}^n)$. Furthermore, $f \in L^2(\mathbb{R}^n)$ belongs to $H_A^1(\mathbb{R}^n)$ if $\int_{\mathbb{R}^n} |(i\nabla - A)f|^2 < \infty$ for some non-zero $A(x)$. Both H^1 and H_A^1 are Hilbert spaces with norms $\|f\|_{H^1}^2 = \int_{\mathbb{R}^n} |f|^2 + \int_{\mathbb{R}^n} |\nabla f|^2$ and $\|f\|_{H_A^1}^2 = \int_{\mathbb{R}^n} |f|^2 + \int_{\mathbb{R}^n} |(i\nabla - A)f|^2$ respectively. For a vector u , if each component of u , $(u)_l$, $l = 1, 2, 3$, belongs to L^p for some $p \in [1, \infty]$, we write $u \in (L^p)^3$.

For the proofs set forth in this article, some different notions of convergence will be used. A sequence $\{\psi_k\} \subset L^p(\mathbb{R}^n)$ is said to converge (in L^p -norm) to $\psi \in L^p(\mathbb{R}^n)$ if and only if

$\int_{\mathbb{R}^n} |\psi_k - \psi|^p \rightarrow 0$, and we write $\psi_k \rightarrow \psi$. Moreover, denote the inner product of a Hilbert space H by $(\cdot, \cdot)_H$. A sequence $\{\psi_k\} \subset H$ is then said to converge weakly to $\psi \in H$ if and only if $(\psi_k, \phi)_H \rightarrow (\psi, \phi)_H$ for all $\phi \in H$, and we write $\psi_k \rightharpoonup \psi$. The inner product of $H^1(\mathbb{R}^n)$ is given by $(\psi, \phi)_{H^1(\mathbb{R}^n)} = \int_{\mathbb{R}^n} \bar{\psi} \phi + \int_{\mathbb{R}^n} \bar{\nabla \psi} \cdot \nabla \phi$. In particular, weak convergence on $H^1(\mathbb{R}^n)$ implies weak convergence in the $L^2(\mathbb{R}^n)$ sense, i.e., $(\psi_k, \phi)_{L^2(\mathbb{R}^n)} = \int_{\mathbb{R}^n} \bar{\psi}_k \phi \rightarrow \int_{\mathbb{R}^n} \bar{\psi} \phi = (\psi, \phi)_{L^2(\mathbb{R}^n)}$.

Also note that a function (or functional) $f : D \rightarrow \mathbb{R}$ is convex on D if for $x_1, x_2 \in D$ and $0 \leq \lambda \leq 1$, we have $f(\lambda x_1 + (1 - \lambda)x_2) \leq \lambda f(x_1) + (1 - \lambda)f(x_2)$.

Now, let ψ denote the wavefunction describing the system. For simplicity, spin will not be treated. Henceforth, assume that $\psi(x_1, \dots, x_N)$ is antisymmetric in its coordinates x_i and belongs to

$$W_N = \{\psi \in H^1(\mathbb{R}^{3N}) \mid \|\psi\|_{L^2(\mathbb{R}^{3N})} = 1\}. \quad (2)$$

Assume $A^k \in L^\infty(\mathbb{R}^3)$, $k = 1, 2, 3$, and $v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$, and define the ground-state energy

$$e_0(v, A) = \inf \{E_{v,A}(\psi) \mid \psi \in W_N\}, \quad (3)$$

where $E_{v,A}(\psi)$ is a functional on W_N given by

$$E_{v,A}(\psi) = \sum_k \left(\int_{\mathbb{R}^{3N}} |(i\nabla_k - A(x_k))\psi|^2 + \int_{\mathbb{R}^{3N}} |\psi|^2 v(x_k) \right) + \sum_{k < l} \int_{\mathbb{R}^{3N}} |\psi|^2 |x_k - x_l|^{-1}. \quad (4)$$

We shall interpret the inner-product $(\psi, H(v, A)\psi)_{L^2}$ as the number $E_{v,A}(\psi)$, which is well-defined for $\psi \in W_N$.

For $\psi \in W_N$, define the particle density and the paramagnetic current density to be, respectively,

$$\begin{aligned} \rho_\psi(x) &= N \int_{\mathbb{R}^{3(N-1)}} |\psi(x, x_2, \dots, x_N)|^2 dx_2 \dots dx_N, \\ j_\psi^p(x) &= N \operatorname{Im} \int_{\mathbb{R}^{3(N-1)}} \bar{\psi}(x, x_2, \dots, x_N) \nabla_x \psi(x, x_2, \dots, x_N) dx_2 \dots dx_N. \end{aligned} \quad (5)$$

Let $H(v, A)$ for the special case $v = 0$ and $A = 0$ be denoted H_0 , that is,

$$H_0 = - \sum_{k=1}^N \Delta_k + \sum_{1 \leq k < l \leq N} |x_k - x_l|^{-1},$$

and set

$$(\psi, H_0\psi)_{L^2} = \sum_k \int_{\mathbb{R}^{3N}} |\nabla_k \psi|^2 dx_1 \dots dx_N + \sum_{k < l} \int_{\mathbb{R}^{3N}} |\psi|^2 |x_k - x_l|^{-1} dx_1 \dots dx_N, \quad (6)$$

for $\psi \in W_N$, even though $H_0\psi \notin L^2$. The kinetic energy of ψ , denoted $T(\psi)$, and the exchange-correlation energy, denoted $E_{xc}(\psi)$, are given by, respectively,

$$T(\psi) = \sum_{k=1}^N \int_{\mathbb{R}^{3N}} |\nabla_k \psi|^2 dx_1 \dots dx_N,$$

$$E_{xc}(\psi) = (\psi, \sum_{1 \leq k < l \leq N} |x_k - x_l|^{-1} \psi)_{L^2} - \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho_\psi(x) \rho_\psi(y)}{|x - y|} dx dy.$$

Note that (6) can be written as $(\psi, H_0\psi)_{L^2} = T(\psi) + E_{xc}(\psi) + \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho_\psi(x) \rho_\psi(y)}{|x - y|} dx dy$.

To put this work into context, the case $A = 0$ will first be discussed. A particle density ρ is said to be v -representable if there exists a Hamiltonian $H(v)$, with ground-state ψ_0 , such that $\rho = \rho_{\psi_0}$. The Hohenberg-Kohn theorem then states that a v -representable particle density ρ determines the scalar potential $v(x)$ up to a constant [1]. For such densities, we can define

$$F_{HK}(\rho) = (\psi_\rho, H(v_\rho)\psi_\rho)_{L^2} - \int_{\mathbb{R}^3} \rho v_\rho = (\psi_\rho, H_0\psi_\rho)_{L^2},$$

where ψ_ρ is the ground-state of $H(v_\rho)$ and where v_ρ is determined by ρ (according to the Hohenberg-Kohn theorem). This scheme, however, suffers from the fact that the functional $F_{HK}(\rho)$ is not explicitly computable, and that the set of v -representable particle densities is unknown. To remedy this situation, Lieb [8] extended the Hohenberg-Kohn functional $F_{HK}(\rho)$ to $F_{LL}(\rho)$ for $\rho \in I_N$, where

$$F_{LL}(\rho) = \inf\{(\psi, H_0\psi)_{L^2} \mid \psi \in W_N, \rho_\psi = \rho\}, \quad (7)$$

and

$$I_N = \left\{ \rho \mid \rho \geq 0, \rho^{1/2} \in H^1(\mathbb{R}^3), \int_{\mathbb{R}^3} \rho = N \right\}.$$

The ground-state energy, $e_0(v)$, can then be obtained from

$$e_0(v) = \inf \left\{ F_{LL}(\rho) + \int_{\mathbb{R}^3} \rho v \mid \rho \in I_N \right\},$$

which is the so-called Levy-Lieb constrained search formalism [8, 9]. Moreover, Lieb [8] has proved that the functional $F_{LL}(\rho)$ is not convex and that the functional

$$F_c(\rho) = \sup \left\{ e_0(v) - \int_{\mathbb{R}^3} \rho v \mid v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3) \right\},$$

which is convex, equals the convex envelope of $F_{LL}(\rho)$ on $L^1(\mathbb{R}^3) \cap L^3(\mathbb{R}^3)$. Furthermore,

$$e_0(v) = \inf \left\{ F_c(\rho) + \int_{\mathbb{R}^3} \rho v \mid \rho \in L^1(\mathbb{R}^3) \cap L^3(\mathbb{R}^3) \right\}.$$

This is the programme we now wish to undertake for paramagnetic current density functionals.

In the remainder of this paper the system Hamiltonian, $H(v, A)$, will also account for a magnetic field $B(x) \neq 0$. For such a system, both the particle density and the paramagnetic current density are needed to describe the system.

III. PARAMAGNETIC CURRENT DENSITY FUNCTIONALS

We will here begin the pursuit of describing a system of N interacting electrons in terms of density functionals with both ρ and j^p as variables. The Vignale and Rasolt formulation of CDFT uses the paramagnetic current density j^p together with the particle density ρ . The statement in [2] that the potentials v and A are determined by the density pair (ρ, j^p) can be reformulated to correctly state that (ρ, j^p) determines the non-degenerate ground-state wavefunction ψ . This ground-state ψ may be the solution to many different Schrödinger equations of the form $H(v, A)\psi = e_0\psi$, where e_0 is the lowest eigenvalue and ψ assumed to be non-degenerate. Hence, the density pair (ρ, j^p) does not necessarily determine the potentials v and A .

With this correspondence between a density pair (ρ, j^p) and a non-degenerate ground-state ψ , the aim is now to generalize some previous results for particle density functionals, i.e., functionals that only depend on ρ . This generalization will follow Lieb's programme for DFT [8], and will constitute of the following: (i) give mathematical criteria for N -representable density pairs (ρ, j^p) , (ii) extend a universal Hohenberg-Kohn functional, denoted $F_{HK}(\rho, j^p)$, to a Levy-Lieb-type functional, denoted $Q(\rho, j^p)$, which has the N -representable densities as domain, (iii) study the convex envelope of $Q(\rho, j^p)$, and (iv) give upper and lower bounds for both $Q(\rho, j^p)$ and $F(\rho, j^p)$.

A. The Hohenberg-Kohn functional and N -representable densities

The starting point is the following theorem:

Theorem 1 *Assume that $H(v_1, A_1)$ and $H(v_2, A_2)$ have non-degenerate ground-states ψ and ϕ respectively. Then $\rho_\psi = \rho_\phi$ and $j_\psi^p = j_\phi^p$ imply $\psi = \text{const. } \phi$.*

Remarks. (i) For a proof we refer either to [2] or Theorem 9 in [6].

(ii) Note that Theorem 1 differs from the Hohenberg-Kohn theorem [1] since no claim is made that the densities determine the potentials.

Theorem 1 will now be applied. The first issue to address is a Hohenberg-Kohn functional that depends on both the density ρ and the paramagnetic current density j^p .

Definition. A density pair (ρ, j^p) is said to be v -representable if there exists a Hamiltonian $H(v, A)$, with ground-state ψ_0 , such that $\rho = \rho_{\psi_0}$ and $j^p = j_{\psi_0}^p$. This set of densities will be denoted A_N , that is,

$$A_N = \{(\rho, j^p) | \rho = \rho_\psi, j^p = j_\psi^p, \psi \text{ is a non-degenerate ground-state of some } H(v, A)\}.$$

For $(\rho, j^p) \in A_N$, let ψ_{ρ, j^p} denote the non-degenerate ground-state of some $H(v, A)$, which is determined by (ρ, j^p) according to Theorem 1. The Hohenberg-Kohn functional, given by

$$F_{HK}(\rho, j^p) = (\psi_{\rho, j^p}, H_0 \psi_{\rho, j^p})_{L^2},$$

is then well-defined for $(\rho, j^p) \in A_N$. Let the set of those potentials v and A such that $H(v, A)$ has a non-degenerate ground-state be denoted V_N , i.e.,

$$V_N = \{(v, A) | H(v, A) \text{ has a non-degenerate ground-state}\}.$$

For $(v, A) \in V_N$, one has

$$e_0(v, A) = \min \left\{ F_{HK}(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \middle| (\rho, j^p) \in A_N \right\}.$$

This is the so-called variational principle of CDFT.

Theorem 2 *For a given potential pair $(v, A) \in V_N$, the ground state energy functional assumes its minimum value for the true ground state densities if the admissible densities are in A_N , i.e.,*

$$e_0(v, A) = \min \left\{ F_{HK}(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \middle| (\rho, j^p) \in A_N \right\}.$$

Proof. Fix $(v, A) \in V_N$. For any $(\rho, j^p) \in A_N$ there exist potentials \tilde{v} and \tilde{A} such that $H(\tilde{v}, \tilde{A})$ has a non-degenerate ground-state ψ_{ρ, j^p} , and one can define

$$G_{v,A}(\rho, j^p) = (\psi_{\rho, j^p}, H(v, A)\psi_{\rho, j^p})_{L^2}.$$

Since ψ_{ρ, j^p} need not be the ground state of $H(v, A)$, by the variational principle for wave-functions

$$G_{v,A}(\rho, j^p) \geq e_0(v, A).$$

Furthermore, by the fact that $(v, A) \in V_N$, there exists a non-degenerate ground state ψ_0 of $H(v, A)$. Let $\rho_0 = \rho_{\psi_0}$ and $j_0^p = j_{\psi_0}^p$, that is, the corresponding ground state particle and paramagnetic current density, which clearly belong to A_N . For $(\rho_0, j_0^p) \in A_N$ there exists ψ_{ρ_0, j_0^p} that satisfies $\psi_{\rho_0, j_0^p} = \text{const. } \psi_0$, by Theorem 1. Hence

$$G_{v,A}(\rho_0, j_0^p) = (\psi_{\rho_0, j_0^p}, H(v, A)\psi_{\rho_0, j_0^p})_{L^2} = e_0(v, A).$$

One may then conclude that

$$\begin{aligned} e_0(v, A) &= \min \{ G_{v,A}(\rho, j^p) \mid (\rho, j^p) \in A_N \} \\ &= \min \left\{ F_{HK}(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \middle| (\rho, j^p) \in A_N \right\}. \blacksquare \end{aligned}$$

The next step is to define a Levy-Lieb-type functional. This functional will be denoted $Q(\rho, j^p)$ and will depend on density pairs (ρ, j^p) that are said to be N -representable. To that end, first note

Proposition 3 (i) *If $\psi \in W_N$, then $\rho_\psi \in I_N$, $j_\psi^p \in (L^1(\mathbb{R}^3))^3$ and $\int_{\mathbb{R}^3} |j_\psi^p|^2 \rho_\psi^{-1} \leq T(\psi)$, and*
(ii) *the functional $(\rho, j^p) \mapsto \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} < \infty$ is convex.*

Proof. (i) Let $\psi \in W_N$, then by Theorem 1.1 of [8], $\rho_\psi \in I_N$. Furthermore, one has

$$\int_{\mathbb{R}^3} |j_\psi^p|^2 \rho_\psi^{-1} \leq \sum_{k=1}^3 \int_{\mathbb{R}^3} \left(N^2 \int_{\mathbb{R}^{3(N-1)}} |\psi|^2 \int_{\mathbb{R}^{3(N-1)}} |\partial_k \psi|^2 \right) \rho_\psi^{-1} = N \int_{\mathbb{R}^{3N}} |\nabla_1 \psi|^2 = T(\psi).$$

To see that each component of j_ψ^p is in $L^1(\mathbb{R}^3)$, note that

$$\int_{\mathbb{R}^3} \left| \int_{\mathbb{R}^{3(N-1)}} \operatorname{Im}(\bar{\psi} \partial_k \psi) \right| \leq \int_{\mathbb{R}^{3N}} |\bar{\psi} \partial_k \psi| \leq \left(\int_{\mathbb{R}^{3N}} |\psi|^2 \right)^{1/2} \left(\int_{\mathbb{R}^{3N}} |\partial_k \psi|^2 \right)^{1/2} < \infty.$$

To prove (ii), set $\rho = \lambda \rho_1 + (1 - \lambda) \rho_2$ and $j^p = \lambda j_1^p + (1 - \lambda) j_2^p$, where $0 < \lambda < 1$. Since

$$\frac{\rho_2}{\rho_1} |j_1^p|^2 + \frac{\rho_1}{\rho_2} |j_2^p|^2 \geq 2 j_1^p \cdot j_2^p,$$

it follows that

$$\begin{aligned} \rho \left(\lambda \frac{|j_1^p|^2}{\rho_1} + (1 - \lambda) \frac{|j_2^p|^2}{\rho_2} \right) &= \lambda^2 |j_1^p|^2 + \lambda(1 - \lambda) \left(\frac{\rho_2}{\rho_1} |j_1^p|^2 + \frac{\rho_1}{\rho_2} |j_2^p|^2 \right) + (1 - \lambda)^2 |j_2^p|^2 \\ &\geq \lambda^2 |j_1^p|^2 + 2\lambda(1 - \lambda) j_1^p \cdot j_2^p + (1 - \lambda)^2 |j_2^p|^2 = |j^p|^2. \end{aligned}$$

One may then conclude

$$\int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} \leq \lambda \int_{\mathbb{R}^3} |j_1^p|^2 \rho_1^{-1} + (1 - \lambda) \int_{\mathbb{R}^3} |j_2^p|^2 \rho_2^{-1},$$

which shows the convexity. \blacksquare

Motivated by Proposition 3, the set of N -representable density pairs (ρ, j^p) is now defined as follows.

Definition. A density pair (ρ, j^p) is said to be N -representable if $(\rho, j^p) \in Y_N$, where $Y_N = \left\{ (\rho, j^p) \mid \rho \in I_N, j^p \in (L^1(\mathbb{R}^3))^3, \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} < \infty \right\}$.

A convex combination of N -representable densities is also N -representable, but a v -representable density need not be N -representable. To summarize

Proposition 4 (i) *The set Y_N is convex, and*

(ii) $A_N \subsetneq Y_N$.

Remark. The proof of part (ii) will be given after Proposition 8.

Proof of (i). Recall that the set I_N consists of those non-negative densities ρ that satisfy $\rho^{1/2} \in H^1(\mathbb{R}^3)$ and $\int_{\mathbb{R}^3} \rho = N$. Note that the functional $\rho \mapsto \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2$ is convex and that I_N is a convex set [8]. Since by Proposition 3 (ii), $(\rho, j^p) \mapsto \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} < \infty$ is a convex functional, it follows that Y_N is a convex set. ■

Note that for $(\rho, j^p) \in Y_N$, $\int_{\mathbb{R}^3} \rho v$ and $\int_{\mathbb{R}^3} j^p \cdot A$ are finite since $v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $A^k \in L^\infty(\mathbb{R}^3)$. However, if a given A has $A^k \notin L^\infty(\mathbb{R}^3)$ for some k , $\int_{\mathbb{R}^3} j^p \cdot A$ is still finite if $(\rho, j^p) \in Y_A$, where $Y_A = \{(\rho, j^p) \in Y_N | \rho \in L^1(\mathbb{R}^3, |A|^2)\}$. Note that if

$$\psi \in \tilde{W}_{N,A} = \{\psi \in \otimes_{k=1}^N H_A^1(\mathbb{R}^3) | \psi \in W_N, \int_{\mathbb{R}^3} \rho_\psi |A|^2 < \infty\},$$

then $(\rho, j^p) \in Y_A$, and

$$\int_{\mathbb{R}^3} |(j^p)_k A^k| \leq \left(\int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} \right)^{1/2} \left(\int_{\mathbb{R}^3} \rho |A|^2 \right)^{1/2} < \infty. \quad (8)$$

The proof of (8) follows directly from $\int_{\mathbb{R}^3} |(j^p)_k A^k| = \int_{\mathbb{R}^3} |(j^p)_k \rho^{-1/2}| |\rho^{1/2} A^k|$ and using Schwarz's inequality.

B. The Levy-Lieb-type functional $Q(\rho, j^p)$

We now turn to finding an extension of the functional $F_{HK}(\rho, j^p)$. A Levy-Lieb-type functional, denoted $Q(\rho, j^p)$, will be introduced (cf. [8] and [9]) and proven to satisfy $Q(\rho, j^p) = F_{HK}(\rho, j^p)$ for $(\rho, j^p) \in A_N$. The domain of $Q(\rho, j^p)$ will consist of those ρ and j^p that are elements of Y_N .

Definition. For $(\rho, j^p) \in Y_N$, we define a Levy-Lieb-type functional

$$Q(\rho, j^p) = \inf \{(\psi, H_0 \psi)_{L^2} | \psi \in W_N, \psi \mapsto (\rho, j^p)\},$$

where $\psi \mapsto (\rho, j^p)$ means that $\rho_\psi = \rho, j_\psi^p = j^p$.

Note that $Q(\rho, j^p)$ is the generalization of $F_{LL}(\rho)$, see (7), when also describing the system with the paramagnetic current density. Theorem 3.3 of [8] states that there exists a $\psi_0 \in H^1(\mathbb{R}^{3N})$ such that $F_{LL}(\rho) = (\psi_0, H_0 \psi_0)_{L^2}$ and $\psi_0 \mapsto \rho$ for $\rho \in I_N$. A similar result is also true for the functional $Q(\rho, j^p)$. Furthermore, on Y_N , $Q(\rho, j^p) \geq \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1}$. The next theorem summarizes the claims made so far about $Q(\rho, j^p)$.

Theorem 5 (i) *There exists a ψ_0 such that $Q(\rho, j^p) = (\psi_0, H_0\psi_0)_{L^2}$ and $\psi_0 \mapsto (\rho, j^p)$,*
(ii) *$Q(\rho, j^p)$ is the proper extension of $F_{HK}(\rho, j^p)$ from A_N to Y_N in the sense that for $(\rho, j^p) \in A_N$, $Q(\rho, j^p) = F_{HK}(\rho, j^p)$, and*
(iii) *$\int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} \leq Q(\rho, j^p)$ on Y_N .*

Proof. (i) Let $\{\psi^k\}_{k=1}^\infty$ be a minimizing sequence, that is $\lim_k (\psi^k, H_0\psi^k)_{L^2} = Q(\rho, j^p)$ and $\psi^k \mapsto (\rho, j^p)$ for all k . From [8] (Theorem 3.3), $\psi^k \rightharpoonup \psi_0$ in $H^1(\mathbb{R}^{3N})$ and $\psi^k \rightarrow \psi_0$ in $L^2(\mathbb{R}^{3N})$ for some H^1 -function ψ_0 (after passing to a subsequence, which we for simplicity continue to denote ψ^k). Then by Theorem 1.3 and Theorem 3.3 in [8], $\rho_{\psi_0} = \rho$. Since taking weak limits, one has $\lim_k (\psi^k, H_0\psi^k)_{L^2} \geq (\psi_0, H_0\psi_0)_{L^2}$. It remains to show that $\psi_0 \mapsto j^p$ a.e.

Let $g(x) = \chi_M(x)$ be the characteristic function of any (measurable) set $M \subset \mathbb{R}^3$ and let $(u)_l$ denote the l :th component of the vector u . Now, using the weak convergence of $\{\psi^k\}_{k=1}^\infty$ in $H^1(\mathbb{R}^{3N})$ and the norm-convergence in $L^2(\mathbb{R}^{3N})$, we have for $l = 1, 2, 3$,

$$\begin{aligned} \lim_{k \rightarrow \infty} \int_{\mathbb{R}^3} (j_{\psi^k}^p)_l g &= \lim_{k \rightarrow \infty} N \operatorname{Im} \int_{\mathbb{R}^3} \int_{\mathbb{R}^{3(N-1)}} \overline{\psi^k} (\partial_l \psi^k) g \\ &= \lim_{k \rightarrow \infty} N \operatorname{Im} \left(\int_{\mathbb{R}^{3N}} (\overline{\psi^k} - \overline{\psi_0}) (\partial_l \psi^k) g + \int_{\mathbb{R}^{3N}} \overline{\psi_0} (\partial_l \psi^k) g \right) = \int_{\mathbb{R}^3} (j_{\psi_0}^p)_l g. \end{aligned}$$

This gives $j_{\psi_0}^p(x) = j_{\psi^k}^p(x) = j^p(x)$ a.e.

(ii) Fix $(\rho, j^p) \in A_N$ and let ψ_0 be as in part (i). The claim in (ii) will be shown by demonstrating that $(\psi_0, H_0\psi_0)_{L^2} = (\psi_{\rho, j^p}, H_0\psi_{\rho, j^p})_{L^2}$, where ψ_{ρ, j^p} satisfies $F_{HK}(\rho, j^p) = (\psi_{\rho, j^p}, H_0\psi_{\rho, j^p})_{L^2}$. Now, since $\psi_{\rho, j^p} \in W_N$ and $\psi_{\rho, j^p} \mapsto (\rho, j^p)$,

$$(\psi_0, H_0\psi_0)_{L^2} = Q(\rho, j^p) \leq (\psi_{\rho, j^p}, H_0\psi_{\rho, j^p})_{L^2}.$$

On the other hand, since ψ_{ρ, j^p} is the ground state of some Hamiltonian $H(v, A)$,

$$\begin{aligned} e_0(v, A) &= (\psi_{\rho, j^p}, H(v, A)\psi_{\rho, j^p})_{L^2} = (\psi_{\rho, j^p}, H_0\psi_{\rho, j^p})_{L^2} + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \\ &\leq (\psi_0, H(v, A)\psi_0)_{L^2} = (\psi_0, H_0\psi_0)_{L^2} + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2), \end{aligned}$$

and $(\psi_0, H_0\psi_0)_{L^2} \geq (\psi_{\rho, j^p}, H_0\psi_{\rho, j^p})_{L^2}$.

(iii) Fix $(\rho, j^p) \in Y_N$. Let $\psi \in W_N$ such that $\rho_\psi = \rho$ and $j_\psi^p = j^p$. By Proposition 3 (i), $\int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} \leq T(\psi)$. We then have

$$\begin{aligned} \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} &\leq \inf \left\{ T(\psi) \mid \psi \in W_N, \psi \mapsto (\rho, j^p) \right\} \\ &\leq \inf \{ (\psi, H_0\psi)_{L^2} \mid \psi \in W_N, \psi \mapsto (\rho, j^p) \} = Q(\rho, j^p). \blacksquare \end{aligned}$$

The situation is now as follows. The set of v -representable density pairs, A_N , is a proper subset of the N -representable density pairs, Y_N . The Hohenberg-Kohn functional F_{HK} , defined on A_N , has been extended to the Levy-Lieb-type functional $Q(\rho, j^p)$, which is defined on Y_N . Combining the variational principle of CDFT with Theorem 5, one has for $(v, A) \in V_N$,

$$e_0(v, A) = \min \left\{ Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \middle| (\rho, j^p) \in A_N \right\}.$$

The admissible set A_N over which the minimization is performed can be exchanged by Y_N if the minimum is replaced by infimum. Note that $Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2)$ remains finite since we require $v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $A^k \in L^\infty(\mathbb{R}^3)$.

Theorem 6 For $v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $A^k \in L^\infty(\mathbb{R}^3)$,

$$e_0(v, A) = \inf \left\{ Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \middle| (\rho, j^p) \in Y_N \right\}.$$

Proof. Fix $(\rho, j^p) \in Y_N$. Then

$$\begin{aligned} e_0(v, A) &= \inf \left\{ (\psi, H(v, A)\psi)_{L^2} \middle| \psi \in W_N \right\} \\ &\leq \inf \left\{ (\psi, H(v, A)\psi)_{L^2} \middle| \psi \in W_N, \rho_\psi = \rho, j_\psi^p = j^p \right\} \\ &= Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2). \end{aligned}$$

Since $(\rho, j^p) \in Y_N$ was arbitrary, we have

$$e_0(v, A) \leq \inf \left\{ Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \middle| (\rho, j^p) \in Y_N \right\}.$$

For the reverse inequality, let $\{\psi_k\}_{k=1}^\infty \subset W_N$ be a minimizing sequence for $e_0(v, A)$, i.e., $e_0(v, A) + \frac{1}{k} > (\psi_k, H(v, A)\psi_k)_{L^2}$. Put $\rho_k = \rho_{\psi_k}$ and $j_k^p = j_{\psi_k}^p$, then

$$\begin{aligned} e_0(v, A) + \frac{1}{k} &> (\psi_k, H_0\psi_k)_{L^2} + 2 \int_{\mathbb{R}^3} j_k^p \cdot A + \int_{\mathbb{R}^3} \rho_k(v + |A|^2) \\ &\geq Q(\rho_k, j_k^p) + 2 \int_{\mathbb{R}^3} j_k^p \cdot A + \int_{\mathbb{R}^3} \rho_k(v + |A|^2) \\ &\geq \inf \left\{ Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \middle| (\rho, j^p) \in Y_N \right\}. \blacksquare \end{aligned}$$

The admissible set over which the minimization is performed can be extended even further. First a definition.

Definition. $X = \{(\rho, j^p) | \rho \in L^1(\mathbb{R}^3) \cap L^3(\mathbb{R}^3), j^p \in (L^1(\mathbb{R}^3))^3\}$.

Next, for $(\rho, j^p) \in X$, define a functional $\tilde{Q}(\rho, j^p)$ given by

$$\begin{aligned}\tilde{Q}(\rho, j^p) &= Q(\rho, j^p) \text{ if } (\rho, j^p) \in Y_N, \\ &= \infty, \text{ otherwise.}\end{aligned}$$

The energy $e_0(v, A)$ can be computed using \tilde{Q} on X . This is implied by the following argument. Let

$$\tilde{e}(v, A) = \inf \left\{ \tilde{Q}(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \mid (\rho, j^p) \in X \right\}.$$

One directly has $\tilde{e}(v, A) \leq e_0(v, A)$, since $\tilde{Q}(\rho, j^p) = Q(\rho, j^p)$ if $(\rho, j^p) \in Y_N$. On the other hand, since $\tilde{Q}(\rho, j^p) = \infty$ if $(\rho, j^p) \notin Y_N$, we have $\tilde{e}(v, A) = e_0(v, A)$. Thus

Theorem 7 For $v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $A^k \in L^\infty(\mathbb{R}^3)$,

$$e_0(v, A) = \inf \left\{ \tilde{Q}(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \mid (\rho, j^p) \in X \right\}.$$

C. Convex envelope of $Q(\rho, j^p)$

So far the variational principle of CDFT has been replaced by the following optimization problem

$$e_0(v, A) = \inf \left\{ Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \mid (\rho, j^p) \in Y_N \right\}.$$

Note that $Q(\rho, j^p)$ could be exchanged by $\tilde{Q}(\rho, j^p)$ and the admissible set Y_N extended to X . However, just as Lieb has demonstrated that $F_{LL}(\rho)$ is not convex (Theorem 3.4 in [8]), the same is also true about $Q(\rho, j^p)$. A proof of this fact as well as a proof of (ii) in Proposition 4 now follows.

Proposition 8 $Q(\rho, j^p)$ is not a convex functional.

Proof of Proposition 8 and Proposition 4 (ii). Choose $v(x)$ as in the proof of Theorem 3.4 in [8] such that it has $M = 2L + 1$ ground-states ψ_k . Set $\rho_k = \rho_{\psi_k}$ and $j_k^p = j_{\psi_k}^p$ for $k = 1, 2, \dots, M$ and note that for all k ,

$$e_0(v, 0) = Q(\rho_k, j_k^p) + \int_{\mathbb{R}^3} \rho_k v. \quad (9)$$

Let $\tilde{\rho} = \frac{1}{M} \sum_{k=1}^M \rho_k$ and $\tilde{j}^p = \frac{1}{M} \sum_{k=1}^M j_k^p$. By definition, $F_{LL}(\tilde{\rho}) \leq Q(\tilde{\rho}, \tilde{j}^p)$. One has

$$e_0(v, 0) < F_{LL}(\tilde{\rho}) + \int_{\mathbb{R}^3} \tilde{\rho} v \leq Q(\tilde{\rho}, \tilde{j}^p) + \int_{\mathbb{R}^3} \tilde{\rho} v,$$

where the first strict inequality follows by Theorem 3.4 of [8] ($\tilde{\rho}$ cannot be a ground-state density of this $v(x)$). Using (9), we obtain

$$\frac{1}{M} \sum_{k=1}^M Q(\rho_k, j_k^p) < Q(\tilde{\rho}, \tilde{j}^p),$$

which shows that $Q(\rho, j^p)$ is not convex.

For the proof of part (ii) in Proposition 4, assume that $\tilde{\rho}$ and \tilde{j}^p are the ground-state densities of some other potential pair (\tilde{v}, \tilde{A}) , then

$$\begin{aligned} e_0(\tilde{v}, \tilde{A}) &= Q(\tilde{\rho}, \tilde{j}^p) + 2 \int_{\mathbb{R}^3} \tilde{j}^p \cdot \tilde{A} + \int_{\mathbb{R}^3} \tilde{\rho}(\tilde{v} + |\tilde{A}|^2) \\ &> \frac{1}{M} \sum_{k=1}^M \left(Q(\rho_k, j_k^p) + 2 \int_{\mathbb{R}^3} j_k^p \cdot \tilde{A} + \int_{\mathbb{R}^3} \rho_k(\tilde{v} + |\tilde{A}|^2) \right). \end{aligned}$$

This gives that for at least one k ,

$$e_0(\tilde{v}, \tilde{A}) > Q(\rho_k, j_k^p) + 2 \int_{\mathbb{R}^3} j_k^p \cdot \tilde{A} + \int_{\mathbb{R}^3} \rho_k(\tilde{v} + |\tilde{A}|^2).$$

But this is a contradiction and hence $A_N \subsetneq Y_N$. ■

The next step will be to obtain a convex and universal density functional, denoted $F(\rho, j^p)$. For that purpose the Legendre transform will be used. The functional $F(\rho, j^p)$ will be defined on the whole space X . (Recall that X is the space of those (ρ, j^p) such that $\rho \in L^1(\mathbb{R}^3) \cap L^3(\mathbb{R}^3)$ and $j^p \in (L^1(\mathbb{R}^3))^3$.)

Definition. The convex functional $F(\rho, j^p)$, defined on X , is given by

$$F(\rho, j^p) = \sup \left\{ e_0(v, A) - 2 \int_{\mathbb{R}^3} j^p \cdot A - \int_{\mathbb{R}^3} \rho(v + |A|^2) \mid v \in L^{3/2} + L^\infty, A^k \in L^\infty \right\}.$$

Remarks. (i) Since F is the supremum over v and A of linear functionals in ρ and j^p , it is convex.

(ii) Furthermore, from the fact that $e_0(v, A) - 2 \int_{\mathbb{R}^3} j^p \cdot A - \int_{\mathbb{R}^3} \rho(v + |A|^2) \leq Q(\rho, j^p)$ for $(\rho, j^p) \in Y_N$, it follows that $F(\rho, j^p) \leq Q(\rho, j^p)$ for all $(\rho, j^p) \in Y_N$.

The functional $F(\rho, j^p)$ can be used to compute the ground-state energy, which follows from a direct generalization of Lieb's proof for the functional

$$\sup \left\{ e_0(v, 0) - \int_{\mathbb{R}^3} \rho v \mid v \in L^{3/2} + L^\infty \right\}.$$

One may minimize $F(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2)$ on either Y_N or X .

Theorem 9

$$\begin{aligned} e_0(v, A) &= \inf \left\{ F(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \mid (\rho, j^p) \in X \right\} \\ &= \inf \left\{ F(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \mid (\rho, j^p) \in Y_N \right\}. \end{aligned}$$

Proof. Denote the first expression of $e_0(v, A)$ as $M^-(v, A)$ and the second one as $M^+(v, A)$. Note that $M^-(v, A) \leq M^+(v, A)$. Now, fix $v_0 \in L^{3/2} + L^\infty$ and $A_0^k \in L^\infty$. By the definition of $F(\rho, j^p)$, we have for $(\rho, j^p) \in X$,

$$F(\rho, j^p) \geq e_0(v_0, A_0) - 2 \int_{\mathbb{R}^3} j^p \cdot A_0 - \int_{\mathbb{R}^3} \rho(v_0 + |A_0|^2) = F_0(\rho, j^p),$$

where the last equality is a definition. Thus

$$\begin{aligned} M^-(v_0, A_0) &\geq \inf \left\{ F_0(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A_0 + \int_{\mathbb{R}^3} \rho(v_0 + |A_0|^2) \mid (\rho, j^p) \in X \right\} \\ &= e_0(v_0, A_0). \end{aligned}$$

But since $v_0 \in L^{3/2} + L^\infty$ and $A_0^k \in L^\infty$ was arbitrary, we obtain $M^-(v, A) \geq e_0(v, A)$.

On the other hand, for $(\rho, j^p) \in Y_N$ we have that $F(\rho, j^p) \leq Q(\rho, j^p)$, and consequently

$$M^+(v, A) \leq \inf \left\{ Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \mid (\rho, j^p) \in Y_N \right\} = e_0(v, A).$$

Thus $e_0(v, A) \leq M^-(v, A) \leq M^+(v, A) \leq e_0(v, A)$. ■

As the reader may recall, $Q(\rho, j^p) = F_{HK}(\rho, j^p)$ on A_N . In fact, $F(\rho, j^p) = Q(\rho, j^p) = F_{HK}(\rho, j^p)$ on A_N , since

Proposition 10 *If $(\rho, j^p) \in A_N$, $F(\rho, j^p) = Q(\rho, j^p)$.*

Proof. Assume $(\rho, j^p) \in A_N$, then $F_{HK}(\rho, j^p) = Q(\rho, j^p)$. For some v and A ,

$$e_0(v, A) = Q(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2).$$

Conversely, using Theorem 9,

$$e_0(v, A) \leq F(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2).$$

Thus $F(\rho, j^p) \geq Q(\rho, j^p)$. However, since the reverse inequality also holds, $F(\rho, j^p) = Q(\rho, j^p)$. ■

The main result of this section is the following:

Theorem 11 *F is the convex envelope of Q .*

Before proving Theorem 11, some preparation is required. First define

Definitions. (i) A functional, f , is weakly lower semi continuous (weakly l.s.c.) if

$$f(\phi) \leq \liminf_{k \rightarrow \infty} f(\phi_k)$$

when $\{\phi_k\}$ converges weakly to ϕ .

(ii) Let Z be a normed space and let $f : D \rightarrow \mathbb{R}$, where $D \subset Z$, and set

$$\Lambda_{f,D} = \{g \mid g \text{ is weakly l.s.c. and convex, and } g(\phi) \leq f(\phi) \text{ for all } \phi \in D\}.$$

The convex envelope on Z of the functional f is then defined to be

$$\text{CE } f(\phi) = \sup\{g(\phi) \mid g \in \Lambda_{f,D}\}.$$

(iii) If Z is a normed space we let Z^* denote the dual space of Z , which is the space of all bounded linear functionals on Z . Moreover, the dual pairing between an element $z \in Z$ and $z^* \in Z^*$ will be denoted $\langle z, z^* \rangle_{Z, Z^*}$. If $Z = L^p(\mathbb{R}^3)$, then $Z^* = L^q(\mathbb{R}^3)$ with $1/p + 1/q = 1$, and $\langle z, z^* \rangle_{Z, Z^*} = \int_{\mathbb{R}^3} z(x) z^*(x) dx$. In particular,

$$X^* = \{(v', A') \mid v' \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3), A' \in (L^\infty(\mathbb{R}^3))^3\}.$$

Proposition 12 F is weakly lower semi continuous.

Proof. The proof of this fact is standard, but is included for the sake of completeness. Since F is convex, it suffices to show that F is lower semi continuous in norm. For any $\lambda \in \mathbb{R}$, define the set

$$\begin{aligned} K_\lambda &= \{(\rho, j^p) | F(\rho, j^p) \leq \lambda\} \\ &= \left\{(\rho, j^p) \left| e_0(v, A) - 2 \int_{\mathbb{R}^3} j^p \cdot A - \int_{\mathbb{R}^3} \rho (v + |A|^2) \leq \lambda, \forall (v, A) \in X^* \right. \right\}. \end{aligned}$$

Now, assume that $\{\rho_n, j_n^p\} \subset K_\lambda$ and that $\rho_n \rightarrow \rho$ in L^1 - and L^3 -norm and that each component of j_n^p converges to the respective component of j^p in L^1 -norm. Then for each $v \in L^{3/2} + L^\infty$ and $A^k \in L^\infty$,

$$\begin{aligned} \lambda &\geq \lim_n \left(e_0(v, A) - 2 \int_{\mathbb{R}^3} j_n^p \cdot A - \int_{\mathbb{R}^3} \rho_n (v + |A|^2) \right) \\ &= e_0(v, A) - 2 \int_{\mathbb{R}^3} j^p \cdot A - \int_{\mathbb{R}^3} \rho (v + |A|^2), \end{aligned}$$

which follows from the fact that norm convergence implies weak convergence. This shows that K_λ is norm closed and that F is norm l.s.c. ■

Now, let f be a convex functional defined on a convex subset $D \subset Z$ of a normed space Z . Then the Legendre transform of f , denoted f^* , defined on the set

$$D^* = \{z^* \in Z^* | \sup_{z \in D} \{\langle z, z^* \rangle_{Z, Z^*} - f(z)\} < \infty\},$$

is given by

$$f^*(z^*) = \sup\{\langle z, z^* \rangle_{Z, Z^*} - f(z) | z \in D\}.$$

Proof of Theorem 11. Let $f = \text{CE } Q$ and $D = Y_N$. Note that $-\text{CE } Q \leq 0$, since $0 \in \Lambda_{Q, Y_N}$. Then, for $(v', A') \in X^*$,

$$f^*(v', A') = \sup \left\{ \int_{\mathbb{R}^3} \rho v' + \int_{\mathbb{R}^3} j^p \cdot A' - \text{CE } Q(\rho, j^p) \left| (\rho, j^p) \in Y_N \right. \right\}.$$

By the definition of the convex envelope, it follows that $\text{CE } Q(\rho, j^p) \leq Q(\rho, j^p)$ on Y_N . This gives

$$\begin{aligned} f^*(v', A') &= \sup \left\{ -\text{CE } Q(\rho, j^p) + \int_{\mathbb{R}^3} \rho v' + \int_{\mathbb{R}^3} j^p \cdot A' \left| (\rho, j^p) \in Y_N \right. \right\} \\ &\geq -\inf \left\{ Q(\rho, j^p) - \int_{\mathbb{R}^3} \rho v' - \int_{\mathbb{R}^3} j^p \cdot A' \left| (\rho, j^p) \in Y_N \right. \right\} \\ &= -e_0(-v' - |A'/2|^2, -A'/2). \end{aligned}$$

By taking the Legendre transform one more time, one obtains for $(\rho, j^p) \in X$

$$\begin{aligned} (f^*)^*(\rho, j^p) &= \sup \left\{ \int_{\mathbb{R}^3} \rho v' + \int_{\mathbb{R}^3} j^p \cdot A' - f^*(v', A') \mid (v', A') \in X^* \right\} \\ &\leq \sup \left\{ e_0(-v' - |A'/2|^2, -A'/2) + \int_{\mathbb{R}^3} \rho v' + \int_{\mathbb{R}^3} j^p \cdot A' \mid (v', A') \in X^* \right\} \\ &= \sup \left\{ e_0(v, A) - 2 \int_{\mathbb{R}^3} j^p \cdot A - \int_{\mathbb{R}^3} \rho (v + |A|^2) \mid (v, A) \in X^* \right\} = F(\rho, j^p), \end{aligned}$$

where $v = -v' - |A'/2|^2 \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $A = -A'/2 \in (L^\infty(\mathbb{R}^3))^3$. We may then conclude that, for $(\rho, j^p) \in X$,

$$(f^*)^*(\rho, j^p) \leq F(\rho, j^p).$$

Now, from an infinite dimensional extension of Fenchel's theorem it follows that if the original functional is convex and weakly lower semi continuous, then the double Legendre transform of the functional equals the functional itself [8]. Thus for $f = \text{CE } Q$ we obtain

$$\text{CE } Q(\rho, j^p) = f(\rho, j^p) = (f^*)^*(\rho, j^p) \leq F(\rho, j^p).$$

Conversely, since F is convex, weakly lower semi continuous and is bounded above by Q , i.e. $F \in \Lambda_{Q, Y_N}$, we have that

$$F(\rho, j^p) \leq \sup \{ f(\rho, j^p) \mid f \in \Lambda_{Q, Y_N} \} = \text{CE } Q(\rho, j^p).$$

It then follows that for all $(\rho, j^p) \in X$, $\text{CE } Q(\rho, j^p) = F(\rho, j^p)$. ■

Since $F(\rho, j^p)$ is convex, one may seek to obtain a connection between a set of Euler-Lagrange equations and the minimization of $F(\rho, j) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho (v + |A|^2)$ on Y_N . Let Z be a normed space and f a real-valued functional on Z , $f : Z \rightarrow \mathbb{R}$. If f is convex on Z , given $z_0 \in Z$, there exists $z^* \in Z^*$, not necessarily unique, such that

$$f(z) \geq f(z_0) + \langle z - z_0, z^* \rangle_{Z, Z^*}$$

holds for all $z \in Z$. We now introduce the concept of Fréchet differentiability and Fréchet derivative. Let $f : Z \rightarrow \mathbb{R}$ be defined on an open domain $D_f \subset Z$. If, for a fixed $z \in Z$ and for each $h \in Z$, there exists $\delta f(z; h) \in \mathbb{R}$ that is linear and continuous with respect to h such that

$$\lim_{\|h\|_Z \rightarrow 0} \frac{|f(z+h) - f(z) - \delta f(z; h)|}{\|h\|_Z} = 0,$$

then f is said to be Fréchet differentiable at z and $\delta f(z; h)$ is said to be the Fréchet differential of f at z with increment h . The Fréchet differential is unique, and if it exists then

$$\lim_{\alpha \rightarrow 0} \frac{f(z + \alpha h) - f(z)}{\alpha}$$

exists and equals the Fréchet differential. We write $\delta f(z; h) = \langle h, f'(z) \rangle_{Z, Z^*}$ and call f' the Fréchet derivative of f . Note that if $f'(z_0)$ exists, we have for all z

$$f(z) \geq f(z_0) + \langle z - z_0, z^* \rangle_{Z, Z^*},$$

where $z^* = f'(z_0)$ is unique. For a functional $f(z_1, z_2)$, f'_{z_k} will be used to denote partial derivative. We are now ready to formulate and prove

Theorem 13 *Assume that $F'_\rho(\rho_0, j_0^p)$ and $F'_{j^p}(\rho_0, j_0^p)$ exist and $\int_{\mathbb{R}^3} \rho_0 = N$ and that*

$$F'_\rho(\rho_0, j_0^p) + v + |A|^2 + \mu_0 = 0,$$

$$F'_{j^p}(\rho_0, j_0^p) + 2A = 0,$$

a.e. for some $\mu_0 \in \mathbb{R}$, $v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $A \in (L^\infty(\mathbb{R}^3))^3$. Then (ρ_0, j_0^p) minimizes

$$\inf \left\{ F(\rho, j^p) + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \mid (\rho, j^p) \in Y_N \right\}.$$

If in addition, $F(\rho_0, j_0^p) = Q(\rho_0, j_0^p)$, then $(\rho_0, j_0^p) \in A_N$.

Proof. Set $w_1 = -F'_\rho(\rho_0, j_0^p) \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $w_2 = -F'_{j^p}(\rho_0, j_0^p) \in (L^\infty(\mathbb{R}^3))^3$. Since F' exists at (ρ_0, j_0^p) and F is convex, we have for $(\rho, j^p) \in Y_N$,

$$F(\rho, j^p) \geq F(\rho_0, j_0^p) + \int_{\mathbb{R}^3} (j_0^p - j^p) \cdot w_2 + \int_{\mathbb{R}^3} (\rho_0 - \rho) w_1.$$

By assumption, $w_1 = v + |A|^2 + \mu_0$ and $w_2 = 2A$ a.e. Since $\int_{\mathbb{R}^3} \rho_0 = N$,

$$F(\rho, j^p) \geq F(\rho_0, j_0^p) + 2 \int_{\mathbb{R}^3} (j_0^p - j^p) \cdot A + \int_{\mathbb{R}^3} (\rho_0 - \rho)(v + |A|^2) + \mu_0(N - \int_{\mathbb{R}^3} \rho).$$

However, for any $(\rho, j^p) \in Y_N$, $\int_{\mathbb{R}^3} \rho = N$, and hence the conclusion follows.

For the second part, assume $F(\rho_0, j_0^p) = Q(\rho_0, j_0^p)$. Using $Q(\rho, j^p) \geq F(\rho, j^p)$, we obtain

$$\begin{aligned} Q(\rho, j^p) &\geq F(\rho, j^p) \geq F(\rho_0, j_0^p) - \int_{\mathbb{R}^3} (\rho - \rho_0) w_1 - \int_{\mathbb{R}^3} (j^p - j_0^p) \cdot w_2 \\ &= Q(\rho_0, j_0^p) - \int_{\mathbb{R}^3} (\rho - \rho_0) w_1 - \int_{\mathbb{R}^3} (j^p - j_0^p) \cdot w_2. \end{aligned}$$

If we define $\tilde{Q}(\rho, j^p) = Q(\rho_0, j_0^p) - \int_{\mathbb{R}^3} (\rho - \rho_0) w_1 - \int_{\mathbb{R}^3} (j^p - j_0^p) \cdot w_2$, we have $\tilde{Q}(\rho, j^p) \leq Q(\rho, j^p)$.

Now,

$$\begin{aligned} Q(\rho_0, j_0^p) + \int_{\mathbb{R}^3} j_0^p \cdot w_2 + \int_{\mathbb{R}^3} \rho_0 w_1 &= \inf \left\{ \tilde{Q}(\rho, j^p) + \int_{\mathbb{R}^3} j^p \cdot w_2 + \int_{\mathbb{R}^3} \rho w_1 \mid (\rho, j^p) \in Y_N \right\} \\ &\leq e_0 \left(w_1 - \frac{|w_2|^2}{4}, \frac{w_2}{2} \right) \leq Q(\rho_0, j_0^p) + \int_{\mathbb{R}^3} j_0^p \cdot w_2 + \int_{\mathbb{R}^3} \rho_0 w_1. \end{aligned}$$

By setting $w_1 = v + |A|^2 \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $w_2 = 2A \in (L^\infty(\mathbb{R}^3))^3$, it follows that

$$e_0(v, A) = Q(\rho_0, j_0^p) + 2 \int_{\mathbb{R}^3} j_0^p \cdot A + \int_{\mathbb{R}^3} \rho_0 (v + |A|^2).$$

From Theorem 5 (i), we know that there exists a $\psi_0 \in W_N$ such that $Q(\rho_0, j_0^p) = (\psi_0, H_0 \psi_0)_{L^2}$ and $\psi_0 \mapsto (\rho_0, j_0^p)$. But then

$$e_0(v, A) = (\psi_0, H_0 \psi_0)_{L^2} - 2 \int_{\mathbb{R}^3} j_0^p \cdot + \int_{\mathbb{R}^3} \rho_0 (v + |A|^2) = (\psi_0, H(v, A) \psi_0)_{L^2},$$

which shows that $(\rho_0, j_0^p) \in A_N$. ■

The last order of business in this section will be to obtain a lower bound for F on X . The motivation is the following. From Theorem 3.8 in [8], we have

$$\begin{aligned} F_c(\rho) = \text{CE } F_{LL}(\rho) &\geq \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2, \text{ if } \rho \in I_N, \\ &\geq \infty, \text{ otherwise.} \end{aligned}$$

We shall now take convex combinations of the two convex functionals $\rho \mapsto \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2$ and $(\rho, j^p) \mapsto N^2 \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1}$ and use Theorem 11 to obtain

Theorem 14 *Define for $(\rho, j^p) \in X$ and $0 \leq \lambda \leq 1$*

$$\begin{aligned} J_\lambda(\rho, j^p) &= \lambda \int_{\mathbb{R}^3} (\nabla \rho(x)^{1/2})^2 + (1 - \lambda) \int_{\mathbb{R}^3} |j^p(x)|^2 \rho(x)^{-1}, \text{ if } (\rho, j^p) \in Y_N, \\ &= \infty, \text{ otherwise.} \end{aligned}$$

Then $J_\lambda(\rho, j^p) \leq F(\rho, j^p)$ for $(\rho, j^p) \in X$ and $0 \leq \lambda \leq 1$.

Proof. From [8] we have that $\rho \mapsto \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2$ is convex and bounded above by $T(\psi)$ for $\rho_\psi = \rho$, $\psi \in W_N$. From Proposition 3 and Theorem 5, we can then conclude that J_λ is convex and $J_\lambda \leq Q$ on Y_N . We now want to show that J_λ is weakly l.s.c. (since J_λ is convex we will show that it is norm-l.s.c) so we can conclude that $J_\lambda \leq \text{CE } Q = F$.

Let $\rho_n \rightarrow \rho$ in $L^1(\mathbb{R}^3) \cap L^3(\mathbb{R}^3)$ -norm and $j_n^p \rightarrow j^p$ in $(L^1(\mathbb{R}^3))^3$ -norm. We want to show that $C_\lambda = \liminf_{n \rightarrow \infty} J_\lambda(\rho_n, j_n^p) \geq J_\lambda(\rho, j^p)$. If $C_\lambda = \infty$ we are done, so we will assume that $C_\lambda < \infty$.

Note that the case $\lambda = 1$ follows from Theorem 3.8 in [8]. It then suffices to show the result for $\lambda = 0$. By the same argument as in the proof of Theorem 3.8 in [8], assume $\rho \in I_N$. (If $\rho < 0$ on a set of positive measure, then $\rho_n < 0$ and $J_0(\rho_n, j_n^p) = \infty$ for sufficiently large n . Similarly we have that $\int_{\mathbb{R}^3} \rho \neq N$ gives $J_0(\rho_n, j_n^p) = \infty$ for sufficiently large n .) Since $C_0 < \infty$, $(\rho_n, j_n^p) \in Y_N$. Set $g_n = j_n^p / \rho_n^{1/2}$. Then $\{g_n\}$ (or at least a subsequence of $\{g_n\}$) is bounded in $L^2(\mathbb{R}^3)^3$, and by the Banach-Alaoglu theorem there exists a $g \in L^2(\mathbb{R}^3)^3$ and a subsequence $\{g_{n_k}\}$ such that $g_{n_k} \rightharpoonup g$ in $L^2(\mathbb{R}^3)^3$.

The next step is to show that $g = j^p / \rho^{1/2}$ a.e. First note since $\rho_{n_k} \rightarrow \rho \geq 0$ in $L^1(\mathbb{R}^3)$ -norm, there exists a subsequence, which we continue to denote $\{\rho_{n_k}\}$, and a non-negative $F \in L^1(\mathbb{R}^3)$ such that $\rho_{n_k}(x) \leq F(x)$ and $\rho_{n_k}(x) \rightarrow \rho(x)$ a.e. From

$$|\rho_{n_k}(x)^{1/2} - \rho(x)^{1/2}|^2 \leq 2(\rho_{n_k}(x) + \rho(x)) \leq 2(F(x) + \rho(x)),$$

we have by dominated convergence, $\rho_{n_k}^{1/2} \rightarrow \rho^{1/2}$ in $L^2(\mathbb{R}^3)$. Let $(u)_l$ denote the l :th component of the vector u . It then follows that $g_{n_k} \rho^{1/2} \rightarrow j^p$ in $L^1(\mathbb{R}^3)^3$, since for $l = 1, 2, 3$,

$$\begin{aligned} \int_{\mathbb{R}^3} |(g_{n_k})_l \rho^{1/2} - (j^p)_l| &\leq \int_{\mathbb{R}^3} |(g_{n_k})_l \rho^{1/2} - (g_{n_k})_l \rho_{n_k}^{1/2}| + \int_{\mathbb{R}^3} |(g_{n_k})_l \rho_{n_k}^{1/2} - (j^p)_l| \\ &\leq \left(\int_{\mathbb{R}^3} |(g_{n_k})_l|^2 \right)^{1/2} \left(\int_{\mathbb{R}^3} |\rho_{n_k}^{1/2} - \rho^{1/2}|^2 \right)^{1/2} + \int_{\mathbb{R}^3} |(j_{n_k}^p)_l - (j^p)_l| \rightarrow 0, \end{aligned}$$

as $k \rightarrow \infty$, where we used that $\rho_{n_k}^{1/2} \rightarrow \rho^{1/2}$ in $L^2(\mathbb{R}^3)$ and $(j_{n_k}^p)_l \rightarrow (j^p)_l$ in $L^1(\mathbb{R}^3)$ for $l = 1, 2, 3$.

Now, let $M \subset \mathbb{R}^3$ be an arbitrary measurable set. Since $\rho^{1/2} \chi_M \in L^2(\mathbb{R}^3)$ and by the weak convergence of g_{n_k} to g in $L^2(\mathbb{R}^3)^3$, one obtains

$$\lim_{k \rightarrow \infty} \int_{\mathbb{R}^3} (g_{n_k})_l \rho^{1/2} \chi_M = \int_M (g)_l \rho^{1/2}.$$

On the other hand, since $\chi_M \in L^\infty(\mathbb{R}^3)$ and norm-convergence implies weak-convergence, $\lim_{k \rightarrow \infty} \int_{\mathbb{R}^3} (g_{n_k})_l \rho^{1/2} \chi_M = \int_M (j^p)_l$. Then $\int_M (g)_l \rho^{1/2} = \int_M (j^p)_l$, which gives $g = j^p / \rho^{1/2}$ a.e.

Lastly, by the w.l.s.c. of the $L^2(\mathbb{R}^3)$ -norm,

$$\begin{aligned} C_0 &= \liminf_{k \rightarrow \infty} \int_{\mathbb{R}^3} |j_{n_k}^p(x)|^2 \rho_{n_k}(x)^{-1} = \liminf_{k \rightarrow \infty} \|g_{n_k}\|_{L^2(\mathbb{R}^3)}^2 \\ &\geq \|g\|_{L^2(\mathbb{R}^3)}^2 = \int_{\mathbb{R}^3} |j^p(x)|^2 \rho(x)^{-1} = J_0(\rho, j^p). \blacksquare \end{aligned}$$

D. Upper and lower bounds for densities with vanishing vorticity

The last issue to be addressed is when the density pair (ρ, j^p) is restricted to the constraint $\nabla \times (j^p/\rho) = 0$. The quantity $\nabla \times (j^p/\rho)$ is called the vorticity. We shall begin by constructing a determinantal wavefunction that yields a prescribed density pair (ρ, j^p) , i.e., finding a function in W_N that is a determinant and that reproduces a given density pair $(\rho, j^p) \in Y_N$. This can be achieved by a straightforward generalization of Theorem 1.2 of [8] (see also [10] where a determinantal construction is considered without the constraint $\nabla \times (j^p/\rho) = 0$ for $N \neq 3$ but without an explicit upper bound for the kinetic energy). Define, as in ref. [8], a function on the real line given by

$$f(x_1) = \frac{2\pi}{N} \int_{-\infty}^{x_1} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \rho(s, x_2, x_3) ds dx_2 dx_3.$$

Note that $f(-\infty) = 0$, $f(\infty) = 2\pi$ and

$$\frac{df}{dx_1} = \frac{2\pi}{N} \int_{\mathbb{R}^2} \rho(x_1, x_2, x_3) dx_2 dx_3.$$

To obtain a determinant ψ_D that yields a given density pair $(\rho, j^p) \in Y_N$, put

$$\psi_D(x_1, \dots, x_N) = (N!)^{-1/2} \det[\phi_k(x_l)]_{k,l},$$

where, for $k = 0, 1, \dots, N-1$,

$$\phi_k(x) = \left(\frac{\rho(x)}{N} \right)^{1/2} e^{i(kf(x_1) - M(x_1) + S(x))}. \quad (10)$$

Note that $(\phi_k, \phi_l)_{L^2} = \delta_{kl}$. It is immediate that $\rho_{\psi_D} = \sum_{k=0}^{N-1} |\phi_k(x)|^2 = \rho$. Moreover, from the calculation

$$\text{Im}(\overline{\phi_k} \nabla \phi_k) = \frac{\rho}{N} \left(\left(k \frac{df}{dx_1} - \frac{dM}{dx_1} \right) \hat{e}_x + \nabla S \right),$$

it follows that

$$\begin{aligned} j_{\psi_D}^p &= \sum_{k=0}^{N-1} \text{Im}(\overline{\phi_k} \nabla \phi_k) = \rho \nabla S + \left(\frac{\rho}{N} \frac{df}{dx_1} \sum_{k=0}^{N-1} k - \rho \frac{dM}{dx_1} \right) \hat{e}_x \\ &= \rho \nabla S + \rho \left(\frac{1}{2} (N-1) \frac{df}{dx_1} - \frac{dM}{dx_1} \right) \hat{e}_x. \end{aligned} \quad (11)$$

Proposition 15 *Given $(\rho, j^p) \in Y_N$ that fulfils $\nabla \times (j^p/\rho) = 0$, there exists a determinant $\psi_D \in L^2(\mathbb{R}^{3N})$ such that $\|\psi_D\|_{L^2} = 1$ and*

$$T(\psi_D) \leq \left(1 + (4\pi)^2 \frac{(N^2 - 1)}{12}\right) \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2 + \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} < \infty. \quad (12)$$

Proof. Take $\psi_D = (N!)^{-1/2} \det[\phi_k(x_l)]_{k,l}$, with ϕ_k as in (10) for $k = 0, 1, \dots, N-1$. From (11), $j_{\psi_D}^p = j^p$ if S and M are chosen such that $\nabla S = j^p/\rho$ and

$$M(x_1) = \frac{f(x_1)}{N} \sum_{k=0}^{N-1} k = \frac{1}{2}(N-1)f(x_1).$$

We are done if we can show (12). To that end, note that

$$|\nabla \phi_k|^2 = \frac{1}{N} \left((\nabla \rho^{1/2})^2 + \rho \left(\left(k \frac{df}{dx_1} - \frac{dM}{dx_1} \right) \hat{e}_x + \nabla S \right)^2 \right).$$

The kinetic energy of ψ_D satisfies

$$\begin{aligned} T(\psi_D) &= \sum_{k=0}^{N-1} \int_{\mathbb{R}^3} |\nabla \phi_k|^2 \\ &= \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2 + \left(\frac{1}{N} \sum_{k=0}^{N-1} k^2 - \frac{(N-1)^2}{4} \right) \int_{\mathbb{R}^3} \rho \left(\frac{df}{dx_1} \right)^2 + \int_{\mathbb{R}^3} \rho |\nabla S|^2 \\ &= \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2 + \frac{(N^2 - 1)}{12} \int_{\mathbb{R}^3} \rho \left(\frac{df}{dx_1} \right)^2 + \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1}. \end{aligned} \quad (13)$$

For the second term in the r.h.s. of (13), note that

$$\int_{\mathbb{R}^3} \rho(x) \left(\frac{df}{dx_1} \right)^2 dx = \left(\frac{2\pi}{N} \right)^2 \int_{\mathbb{R}} g(x_1)^6 dx_1,$$

where

$$g(x_1)^2 = \int_{\mathbb{R}^2} \rho(x_1, x_2, x_3) dx_2 dx_3.$$

From [8], $g \in H^1(\mathbb{R})$ and moreover

$$g(x_1)^4 \leq 4 \int_{\mathbb{R}} g(x_1)^2 dx_1 \int_{\mathbb{R}} \left| \frac{dg}{dx_1} \right|^2 dx_1 \leq 4N \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2 dx.$$

Thus, (13) now gives

$$T(\psi_D) \leq \left(1 + (4\pi)^2 \frac{(N^2 - 1)}{12}\right) \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2 + \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} < \infty,$$

where all terms are finite since $(\rho, j^p) \in Y_N$. \blacksquare

Remark. Note that for ψ_D chosen as in Proposition 15, the exchange-correlation energy, $E_{xc}(\psi_D)$, does not depend on the paramagnetic current density. This can be seen from

$$\begin{aligned} E_{xc}(\psi_D) &= -\frac{1}{2N^2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho(x)\rho(y)}{|x-y|} \left| \sum_{k=0}^{N-1} e^{ik(f(x_1)-f(y_1))} \right|^2 dx dy \\ &= -\frac{1}{2N} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho(x)\rho(y)}{|x-y|} F_N(f(x_1) - f(y_1)) dx dy, \end{aligned}$$

where $F_N(t)$ is the Fejér kernel, given by $F_N(t) = \sin^2(Nt/2) / (N \sin^2(t/2))$.

Proposition 16 For $(\rho, j^p) \in Y_N$ fulfilling $\nabla \times (j^p/\rho) = 0$, we have

$$Q(\rho, j^p) \leq \left(1 + (4\pi)^2 \frac{(N^2 - 1)}{12}\right) \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2 + \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} + \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho(x)\rho(y)}{|x-y|} dx dy.$$

Proof. First note that

$$(\psi, H_0 \psi) = T(\psi) + E_{xc}(\psi) + \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho_\psi(x)\rho_\psi(y)}{|x-y|} dx dy.$$

Now, given $(\rho, j^p) \in Y_N$ fulfilling $\nabla \times (j^p/\rho) = 0$, there exists, by Proposition 15, a determinantal wavefunction ψ_D such that $\rho_{\psi_D} = \rho$ and $j_{\psi_D}^p = j^p$. We then have

$$\begin{aligned} Q(\rho, j^p) &\leq \left(1 + (4\pi)^2 \frac{(N^2 - 1)}{12}\right) \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2 + \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} + (\psi_D, \sum_{1 \leq k < l \leq N} |x_k - x_l|^{-1} \psi_D) \\ &\leq \left(1 + (4\pi)^2 \frac{(N^2 - 1)}{12}\right) \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2 + \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} + \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho(x)\rho(y)}{|x-y|} dx dy, \end{aligned}$$

where the last inequality follows from the fact that $E_{xc}(\psi_D) \leq 0$, since ψ_D is a determinant. \blacksquare

We conclude this section by applying Proposition 16 and Theorem 14. The following corollary gives both an upper and lower bound for Q and F in terms of $J_0(\rho, j^p) = \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1}$ and $J_1(\rho, j^p) = \int_{\mathbb{R}^3} (\nabla \rho^{1/2})^2$.

Corollary 17 *Let $(\rho, j^p) \in Y_N$ be such that $\nabla \times (j^p/\rho) = 0$. Then for $0 \leq \lambda \leq 1$,*

$$\begin{aligned} \lambda J_0(\rho, j^p) + (1 - \lambda) J_1(\rho, j^p) &= J_\lambda(\rho, j^p) \leq F(\rho, j^p) \leq Q(\rho, j^p) \\ &\leq aN + (b + cN^2) J_1(\rho, j^p) + J_0(\rho, j^p), \end{aligned}$$

where $a = 4/(3\sqrt{3}\pi)$, $b = 1 - (4\pi)^2/12$ and $c = (4\pi)^2/12 + 4/(3\sqrt{3}\pi)$.

Proof. The statement follows directly from Proposition 16 and Theorem 14 and the fact that

$$\begin{aligned} \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho(x)\rho(y)}{|x-y|} dx dy &\leq C_1 \|\rho\|_{L^{6/5}(\mathbb{R}^3)}^2 \leq C_1 N^{3/2} \|\rho\|_{L^3(\mathbb{R}^3)}^{1/2} \\ &\leq C_1 C_2 N^{3/2} J_1(\rho, j^p)^{1/2} \leq \frac{1}{\pi} \frac{4}{3\sqrt{3}} (N + N^2 J_1(\rho, j^p)), \end{aligned}$$

where the Hardy-Littlewood-Sobolev inequality ($C_1 = 2(4/\pi^{1/2})^{2/3}/3$) and Sobolev's inequality for gradients ($C_2 = 2/(3^{1/2}2^{1/3}\pi^{2/3})$) have been used [11]. ■

IV. SUMMARY

This paper has aimed at giving CDFT formulated with the paramagnetic current density a mathematically rigorous foundation. It has focused on defining and investigating density functionals that depend on the particle density and the paramagnetic current density. N -representable density pairs (ρ, j^p) have been defined. A Hohenberg-Kohn functional, $F_{HK}(\rho, j^p)$, has been extended to a Levy-Lieb-type functional, denoted $Q(\rho, j^p)$, with the set of N -representable densities as domain. It has been proven that there exists a wavefunction ψ_0 such that $Q(\rho, j^p) = (\psi_0, H_0 \psi_0)_{L^2}$ and $\rho_\psi = \rho$, $j_\psi^p = j^p$. Moreover, a universal and convex density functional $F(\rho, j^p)$ has been proven to exist such that it equals the convex envelope of $Q(\rho, j^p)$. On the set of v -representable densities, the functionals F_{HK} , Q and F all agree. Furthermore, a connection between the minimization of $F(\rho, j^p)$ and a set of Euler-Lagrange equations has been established.

For N -representable density pairs (ρ, j^p) fulfilling $\nabla \times (j^p/\rho) = 0$, both upper and lower bounds of F and Q in terms of convex functionals that are given explicitly have been obtained.

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- [1] P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (Nov 1964),
<http://link.aps.org/doi/10.1103/PhysRev.136.B864>
- [2] G. Vignale and M. Rasolt, Phys. Rev. Lett. **59**, 2360 (Nov 1987),
<http://link.aps.org/doi/10.1103/PhysRevLett.59.2360>
- [3] G. Diener and M. Rasolt, J. Phys.: Condens. Matter **3**, 9417 (1991)
- [4] X.-Y. Pan and V. Sahni, Int. Jour. Quant. Chem. **110**, 2833 (2010),
<http://dx.doi.org/10.1002/qua.22862>
- [5] K. Capelle and G. Vignale, Phys. Rev. B **65**, 113106 (Feb 2002),
<http://link.aps.org/doi/10.1103/PhysRevB.65.113106>
- [6] A. Laestadius and M. Benedicks, To appear in Int. Jour. Quant. Chem.
- [7] E. I. Tellgren, S. Kvaal, E. Sagvolden, U. Ekström, A. M. Teale, and T. Helgaker, Phys. Rev. A **86**, 062506 (Dec 2012), <http://link.aps.org/doi/10.1103/PhysRevA.86.062506>
- [8] E. H. Lieb, Int. Jour. Quant. Chem. **24**, 243 (1983), <http://dx.doi.org/10.1002/qua.560240302>
- [9] M. Levy, Proc. Natl. Acad. Sci. USA **76**, 6062 (1979)
- [10] E. H. Lieb and R. Schrader, Phys. Rev. A **88**, 032516 (Sep 2013),
<http://link.aps.org/doi/10.1103/PhysRevA.88.032516>
- [11] E. H. Lieb and M. Loss, *Analysis* (American Mathematical Society, Providence, Rhode Island, 2001)